

Phonon-mediated coupling between quantum dots through an off-resonant microcavity

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We present experimental results showing phonon-mediated coupling between two quantum dots embedded inside a photonic crystal microcavity. With only one of the dots being spectrally close to the cavity, we observe both frequency up-conversion and down-conversion of the pump light via a ~ 1.2 THz phonon. We demonstrate this process for both weak and strong regimes of dot-cavity coupling, and provide a simple theoretical model explaining our observations.

I. INTRODUCTION

Phonon-mediated coupling between a self-assembled semiconductor quantum dot (QD) and a semiconductor microcavity is a recently discovered phenomenon unique to solid state cavity quantum electrodynamics [1, 2]. Due to the electron-phonon interaction in the QD, we observe cavity emission when the QD is resonantly excited. Alternatively, we see emission from the QD when the cavity is pumped on resonance. Apart from the fundamental interest in identifying the mechanism behind this unusual off-resonant coupling [3, 4], this effect can be used to probe the coherent interaction of the QD with a strong laser [5] and the cavity-enhanced AC stark shift of a QD [6]. These results demonstrate that the off-resonant cavity constitutes an efficient read-out channel for the QD states.

However, all experiments reported so far in the literature are based on the interaction between a single QD and a cavity. Recently, an experimental study of two spatially separated QDs interacting resonantly in a microcavity has been reported [7], as well as a theoretical analysis [8] of the possible energy transfer mechanisms between QDs in such a cavity. In this work, we show that two spectrally detuned QDs can interact with each other via an off-resonant cavity. More specifically, we observe emission from a spectrally detuned QD when another QD is resonantly excited. Both frequency down-conversion (transfer of photons from a higher energy QD to a lower energy QD) and up-conversion (transfer of photons from a lower energy QD to a higher energy QD) are observed for frequency separation of up to $\sim \pm 1.2$ THz. Such a large energy difference cannot be ascribed to an excited state of the same QD, as opposed to earlier work by Flagg et al [9] which was performed without a cavity and for frequency difference of $\sim \pm 0.2$ THz. Based on our observations, we believe this process is happening between two different QDs, and the coupling between the QDs is enhanced by the presence of the cavity.

II. THEORY

The experimental system we want to model is shown in Fig. 1a. QD1, spectrally detuned from both the cavity and QD2, is resonantly excited with a pump laser. The excitation is transferred to the cavity and QD2 via an incoherent phonon-mediated coupling [10]. We note that, in theory, it is possible to transfer energy directly from QD1 to QD2 via phonons. However, we observe the QD2 emission to be strongly dependent on the QD2-cavity detuning, and hence the presence of a cavity is important for our experiment. In particular, for detunings greater than a few cavity linewidths, the QD2 emission becomes weak and eventually vanishes.

The Master equation to describe the lossy dynamics of the density matrix ρ of a coupled system consisting of two QDs and a cavity is given by

$$\frac{d\rho}{dt} = -i[\mathcal{H}, \rho] + 2\kappa\mathcal{L}[a] + 2\gamma_1\mathcal{L}[\sigma_1] + 2\gamma_2\mathcal{L}[\sigma_2]. \quad (1)$$

Assuming the rotating wave approximation, the Hamiltonian describing the coherent dynamics of the system \mathcal{H} can be written in the interaction picture as

$$\mathcal{H} = \omega_c a^\dagger a + \omega_{d1} \sigma_1^\dagger \sigma_1 + g_1 (a^\dagger \sigma_1 + a \sigma_1^\dagger) + \omega_{d2} \sigma_2^\dagger \sigma_2 + g_2 (a^\dagger \sigma_2 + a \sigma_2^\dagger), \quad (2)$$

while the Lindblad operator modeling the incoherent decay via a collapse operator D is $\mathcal{L}[D] = D\rho D^\dagger - \frac{1}{2}D^\dagger D\rho - \frac{1}{2}\rho D^\dagger D$. Additionally, κ is the cavity field decay rate; γ_1 and γ_2 are the QD dipole decay rates; ω_c , ω_{d1} and ω_{d2} are the resonance frequencies of the cavity, QD1 and QD2; g_1 and g_2 are the coherent interaction strengths between the cavity and the two QDs. The resonant driving of QD1 or QD2 can be described, respectively, by adding the term $\Omega(\sigma_1 + \sigma_1^\dagger)$ or $\Omega(\sigma_2 + \sigma_2^\dagger)$ to the Hamiltonian \mathcal{H} . The driving laser frequency is denoted by ω_l . We model the incoherent phonon-mediated coupling by adding $2\gamma_{r1}\mathcal{L}[a^\dagger \sigma_1]$ and $2\gamma_{r2}\mathcal{L}[a \sigma_2^\dagger]$ to the Master equation. The channel between QD1 and the cavity is then characterized by the values of γ_{r1} and g_1 , while the channel between the cavity and QD2 is characterized by γ_{r2} and g_2 . Fig. 1c shows the numerically simulated power spectral density (PSD) of the QD resonance fluorescence $S(\omega) = \int_{-\infty}^{\infty} \langle a^\dagger(\tau)a(0) \rangle e^{-i\omega\tau} d\tau$ collected through the

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cavity. We use only the cavity operator to calculate the PSD because experimentally most of the collected light is in the cavity mode. For these simulations, we use $\gamma_1/2\pi = \gamma_2/2\pi = 1\text{GHz}$, $\gamma_{r1}/2\pi = 0.5\text{GHz}$, $g_1/2\pi = 20\text{GHz}$, $\kappa/2\pi = 20\text{GHz}$, QD1-cavity detuning $\Delta_1 = 6\kappa$ and QD2-cavity detuning $\Delta_2 = -6\kappa$ and the driving laser strength $\Omega_0/2\pi = 5\text{GHz}$.

We first study the role of γ_{r2} and g_2 in the QD2 emission. Without g_2 , no emission from QD2 is observed; in the presence of g_2 , QD2 emission appears and γ_{r2} enhances it (Fig. 1 c). This shows that coherent coupling between the cavity and QD2 is required to observe this dot to dot coupling. The three peaks observed at the QD1 resonance are the usual Mollow triplet, modified due to the presence of the cavity and phonons [10–12].

Next, we analyze the dependence of the inter-dot coupling on the spectral detuning between the undriven dot and the cavity. In an actual experiment it is very difficult to tune only one QD without affecting the other, as the two QDs are spatially very close to each other. Hence, in the simulation, we changed both QD resonances and kept the cavity resonance fixed. In Fig. 1d we excite QD1, which is spectrally far detuned from the cavity. QD2 is spectrally close to the cavity, and strongly coupled. The resonant excitation of QD1 causes light to be emitted both from the cavity and from QD2. Additionally, we observe anti-crossing between the cavity and QD2 as the frequency of QD2 is tuned. Following this, we excite QD2 resonantly, and observe emission from QD1 (Fig. 1e). We observe an increase in QD1 emission intensity when QD2 is resonant with the cavity. Finally, we calculate the linewidth of QD1 while measuring the emission from QD2 (inset of Fig. 1d), as well as the linewidth of QD2 while measuring the emission from QD1 (inset of Fig. 1e), for a weak excitation laser power ($\Omega_0/2\pi = 1\text{GHz}$). We find that the linewidth of QD1 is 4 GHz and the linewidth of QD2 is 9 GHz. These simulated linewidths are larger than the linewidths one would expect based on the decay rates, i.e. $2(\gamma + \gamma_r)/2\pi = 3\text{GHz}$, and are the result of the cavity (with a linewidth of 40 GHz) assisting in the coupling between QD1 and QD2.

III. TEMPORAL DYNAMICS OF DOT-CAVITY OFF-RESONANT COUPLING

In this section, we describe an experiment to estimate the time required to transfer the energy from a QD to the cavity, when the QD is resonantly excited. This measurement gives a way to estimate the incoherent coupling rate γ_r . The experiments are performed in a helium-flow cryostat at cryogenic temperatures ($\sim 30 - 55\text{K}$) on self-assembled InAs QDs embedded in a GaAs three-hole defect L_3 photonic crystal cavity [13]. The 160nm GaAs membrane used to fabricate the photonic crystal is grown by molecular beam epitaxy on top of a GaAs (100) wafer. The GaAs membrane sits on a 918 nm sacrificial layer of $\text{Al}_{0.8}\text{Ga}_{0.2}\text{As}$. Under the sacrificial layer, a 10-period

distributed Bragg reflector consisting of a quarter-wave AlAs/GaAs stack is used to increase the signal collection into the objective lens. The photonic crystal was fabricated using electron beam lithography, dry plasma etching, and wet etching of the sacrificial layer [13].

We resonantly excite the QD with a laser pulse train consisting of $\sim 40\text{ps}$ wide pulses with a repetition period of 13 ns. A grating filter is used to collect only the off-resonant cavity emission and block all the background light from the excitation laser. The cavity emission signal is then sent to a single photon counter followed by a picosecond time analyzer (PTA). The PTA is triggered by the excitation laser pulse, and the cavity emission is recorded. Fig. 2 shows the pulse shape, as well as the rising and falling edges of the cavity emission for different temperatures. By fitting exponentials to the cavity signal, we estimate the rise and fall times of the cavity emission. The rise time gives an estimate of the excitation transfer time τ_r between the QD and the cavity, and in our system it is on the order of $\sim 500\text{ps}$. From this we use the formula $\gamma_r \bar{n} = 1/\tau_r$ to roughly estimate $\gamma_r/2\pi$ to be 0.25 GHz, assuming the mean phonon number $\bar{n} \approx 8$. The QD in this particular case is blue detuned from the cavity, so to have off-resonant coupling, a phonon needs to be absorbed. The temperature is changed from 40 K to 50 K, corresponding to a change in dot-cavity detuning from 1.8 nm to 2.25 nm and a change in mean phonon number \bar{n} from 7.8 to 8. Similar values of rise time are obtained for several different QDs, both blue and red detuned from the cavity. The fall time corresponds to the cavity life-time, but the measured values of $\sim 850\text{ps}$ are rather large compared to the values previously reported [14]. This might be due to the high density of QDs in the sample, as well as the presence of phonons.

IV. COUPLING BETWEEN TWO QUANTUM DOTS

In this section, we present experimental data showing dot to dot coupling via an off-resonant cavity for two different systems: one with a strongly coupled QD, and the other with a weakly coupled QD. In the first system, we excite QD1 resonantly with a laser, and observe emission both from the off-resonant cavity and QD2 (Fig. 3). Note that QD2 is strongly coupled to the cavity and we observe anti-crossing between the cavity and QD2 in the off-resonant emission when the temperature of the system is changed (inset of Fig. 3). The experimental data match well qualitatively with the theoretical result shown in Fig. 1d. The emission from QD2 diminishes as QD2 is detuned from the cavity, which shows that the coupling between the two dots is enhanced by the presence of the cavity. However, when we scan the pump laser across QD2 and observe QD1 emission in this system, we obtain the cavity linewidth showing the usual cavity to QD1 coupling [15]. This might be due to the high temperature (40-48K) of the system, as will be explained

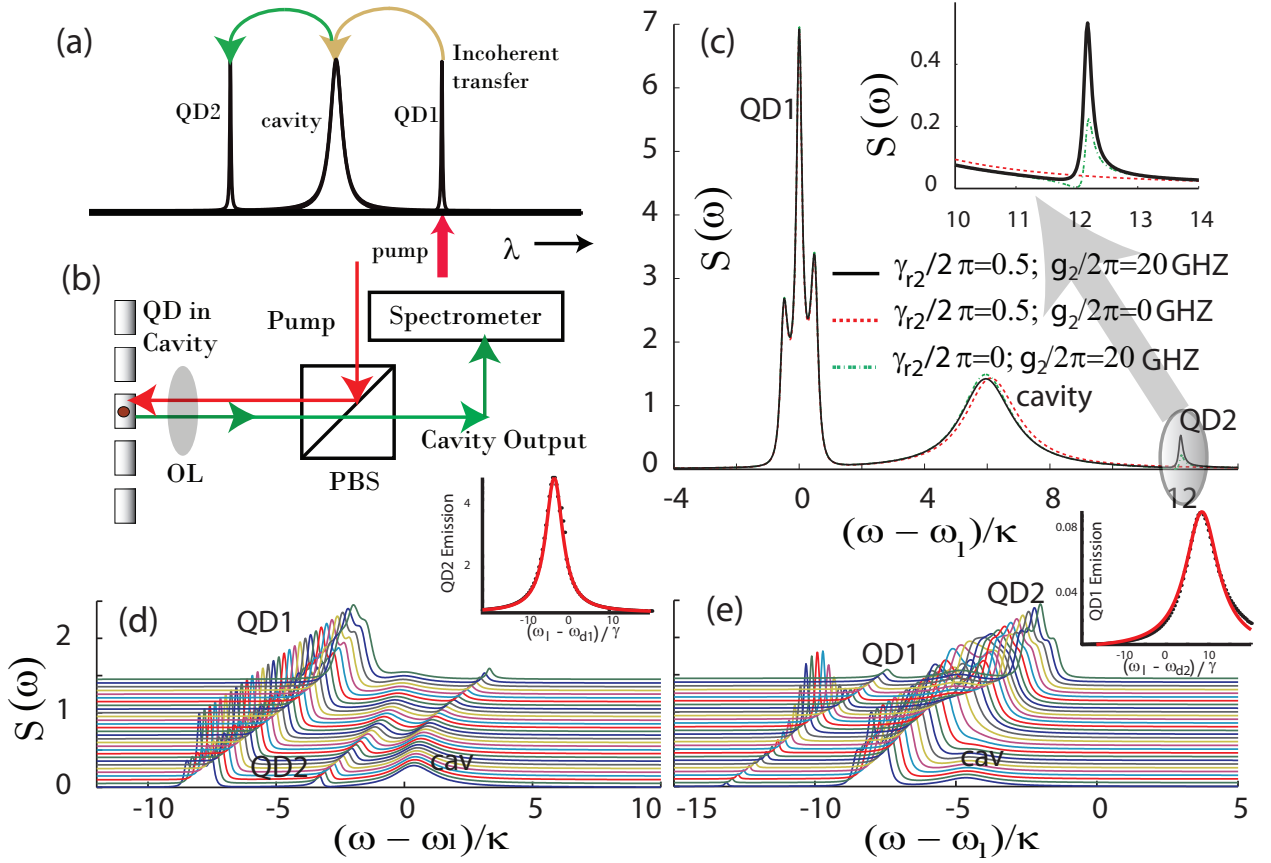


FIG. 1: (color online) (a) Illustration of the dot to dot coupling. QD1 is resonantly driven with a pump laser. The excitation is transferred to QD2 via the cavity. (b) Experimental cross-polarized confocal microscopy setup. PBS: polarizing beam splitter, OL: objective lens. (c) Numerically calculated power spectral density $S(\omega)$ of the QD resonance fluorescence collected through the cavity for different γ_{r2} and g_2 , when QD1 is resonantly excited (i.e., $\omega_l = \omega_{d1}$). Inset shows a zoom-in of the QD2 emission. (d), (e) Calculated $S(\omega)$ when QD1(d) and QD2(e) are resonantly excited, and the dot-cavity detunings are changed. In other words, $\omega_l = \omega_{d1}$ for (d) and $\omega_l = \omega_{d2}$ for (e). Both QD1 and QD2 resonances are shifted. We observe anti-crossing between QD2 and the cavity in the off-resonant emission (d). We also observe an increase in QD1 emission when QD2 is resonant with the cavity (e). For (d) and (e) parameters used for the simulations were: $\gamma_1/2\pi = \gamma_2/2\pi = 1$ GHz, $\gamma_{r1}/2\pi = \gamma_{r2}/2\pi = 0.5$ GHz, $g_1/2\pi = g_2/2\pi = 20$ GHz, $\kappa/2\pi = 20$ GHz, detuning between two dots 5κ , driving laser strength $\Omega_0/2\pi = 5$ GHz.

later in this paper.

In the second system, the QD that is spectrally close to the cavity is only weakly coupled to it. We observe emission from a lower energy QD when a laser resonantly excites a higher energy QD (Fig. 4 blue plot). We also observe up-conversion, i.e., emission from the higher energy QD under excitation of the lower energy QD (Fig. 4 red plot). The energy difference between the two QDs corresponds to a 1.2 THz acoustic phonon. The cavity is at ~ 935 nm, closer to the higher energy QD, although its emission is not distinctly noticeable. The data is taken at 25K. In the inset (replicated in Fig. 5 a,b), we plot the collected emission from a QD and estimate the linewidth of the other (excited) QD by a Lorentzian fit. The higher and lower energy QDs, respectively, have linewidths of ~ 0.03 nm and ~ 0.013 nm. These are comparable to the linewidths of the self-assembled QDs [15], and indicate that the coupling is indeed between two

QDs. The broader linewidth of the higher energy QD is due to the presence of the cavity. Following this, we perform a more accurate measurement of the linewidths of each QD by observing the peak amplitude of the emission from the off-resonant dot as a function of the pump laser wavelength λ_p (Fig. 5 c,d) [15]. From the Lorentzian fit, we estimate that linewidths of the higher and lower energy QDs are, respectively, ~ 0.024 nm and ~ 0.008 nm. The slightly smaller linewidths measured by the latter approach are due to the better spectral resolution offered by this method [15].

Finally, we performed a study of the effects of temperature on the inter-dot coupling. We note that while down-conversion of the pump light is observed at a temperature as low as 10 K, we did not observe any up-conversion at this temperature. This corroborates the fact that the observed dot to dot coupling is phonon-mediated, and at a low temperature up-conversion cannot happen due to

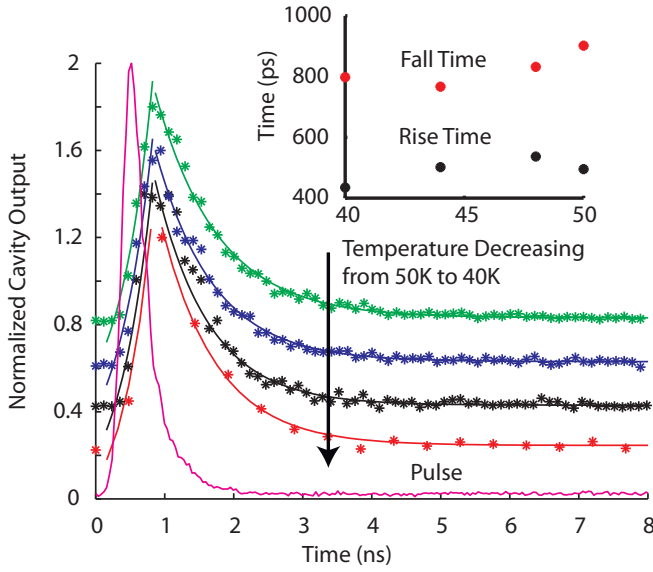


FIG. 2: (color online) Time resolved measurement of the off-resonant cavity emission. The QD is resonantly excited with a 40 ps pulse, and the time-resolved measurement of the off-resonant cavity emission is performed. The inset plots the rise times and fall times of the cavity emission (extracted from the exponential fits) against the system's temperature.

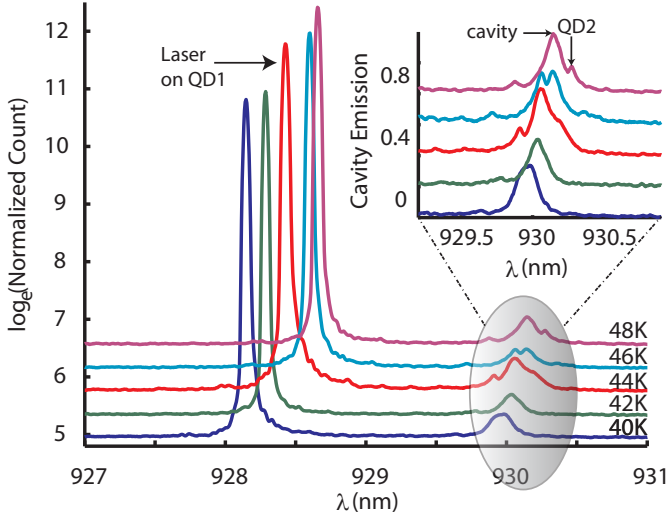


FIG. 3: (color online) Measurement of the emission from the off-resonant cavity and QD2, under resonant excitation of QD1. We observe anti-crossing between QD2 and the cavity when the temperature of the system is changed.

the smaller number of phonons. We first scan the laser across the higher energy QD and observe the off-resonant emission from the lower energy QD; Fig. 6a shows the result of this measurement for a set of different temperatures. Similarly, Fig. 6b shows the data obtained by scanning the laser across the lower energy QD and observing the off-resonant emission from the higher energy

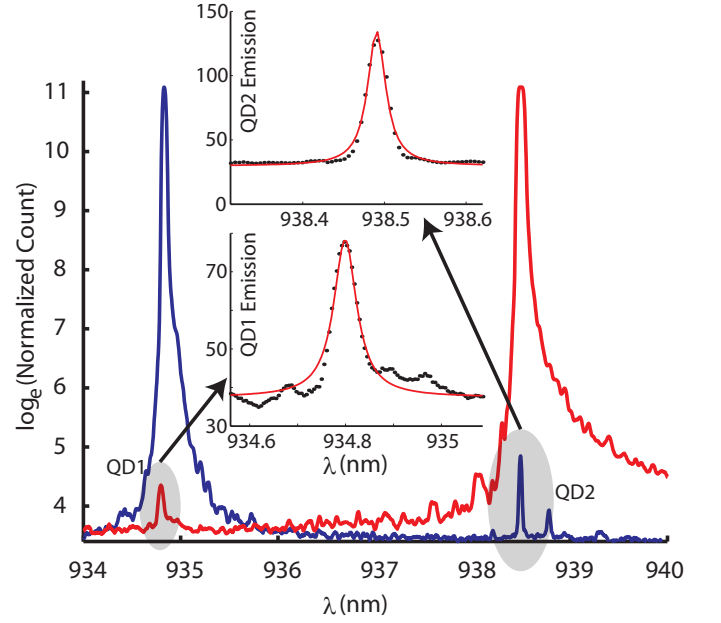


FIG. 4: (color online) Experimental demonstration of the phonon mediated inter-dot coupling: we observe the emission from a lower energy QD, when a higher energy quantum dot is resonantly excited (blue). Similarly, under resonant excitation of a lower energy QD, emission from a higher energy QD is observed (red). Inset zooms into the QD emission. QD linewidths are estimated by fitting Lorentzians. Measured linewidths of the higher and lower energy QDs, respectively, are ~ 0.03 nm and ~ 0.013 nm. The cavity is at ~ 935 nm, close to the higher energy QD.

QD for an assortment of temperatures. It can be seen from the down-conversion plots in Fig. 6a that at lower temperature we observe emission from the lower energy QD only when the pump is within the linewidth of the higher energy QD. However, with increasing temperature we observe emission from the lower energy QD even when the cavity is pumped. When the temperature is raised to ~ 40 K, we observe coupling only from cavity to the lower energy QD, similar to the observations reported previously [2]. We note that for the experimental result (performed at a temperature of ~ 45 K) shown in Figure 3 (for the system only with a strongly coupled dot), we also observe coupling between the cavity and the QD, and not between two QDs. This disappearance of dot to dot down-conversion might be caused by the increase in phonon density and the resulting broadening of the QD lines. In Fig. 6b, we monitor the effects of temperature on the up-conversion. We cannot detect the up-conversion at 10 K, as it only becomes observable at higher temperatures. However, with increasing temperature, the QD lines disappear. This is most likely due to the fact that the QD starts losing confinement with higher temperature. The additional peaks in Fig. 6b show up-conversion of several other QDs.

In summary, we observed phonon mediated inter-dot coupling, both in systems with strongly and weakly cou-

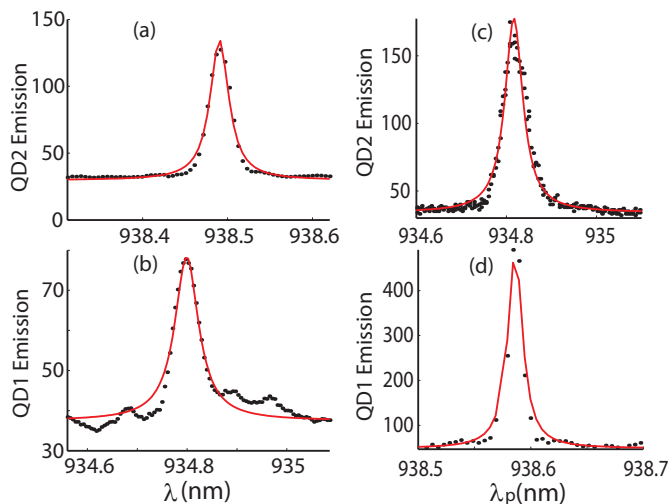


FIG. 5: (color online) Comparison of the linewidths measured in direct off-resonant dot emission (Fig. 4) and from resonant spectroscopy of the QDs: (a),(b) the off-resonant QD emission (same as the inset of Fig. 4). QD linewidths are estimated by fitting Lorentzians. Linewidths of the higher and lower energy QDs, respectively, are ~ 0.03 nm and ~ 0.013 nm. The cavity is at ~ 935 nm, close to the higher energy QD. (c),(d) the off-resonant dot emission as a function of the pump laser wavelength λ_p . In this experiment, a laser is scanned across one QD and emission is collected from the other QD as a function of the laser wavelength, same as in Ref. [15]. By fitting Lorentzians, we estimate the linewidths of the higher and lower energy QDs to be ~ 0.024 nm and ~ 0.008 nm, respectively.

pled QDs. Both frequency up- and down-conversion were reported via a phonon of estimated energy ~ 1.2 THz. Our results indicate that this coupling is enhanced by the presence of the cavity, and that without a cavity spectrally close to one of the QDs this process does not occur.

V. ACKNOWLEDGEMENTS

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- [1] S. Ates et al., *Nature Photonics* **3**, 724 (2009).
 - [2] D. Englund, A. Majumdar, A. Faraon, M. Toishi, N. Stoltz, P. Petroff, and J. Vučković, *Phys. Rev. Lett.* **104**, 073904 (2010).
 - [3] M. Calic, P. Gallo, M. Felici, K. A. Atlasov, B. Dwir, A. Rudra, G. Biasiol, L. Sorba, G. Tarel, V. Savona, et al., *Phys. Rev. Lett.* **106**, 227402 (2011).
 - [4] M. Winger, T. Volz, G. Tarel, S. Portolan, A. Badolato, K. J. Hennessy, E. L. Hu, A. Beveratos, J. Finley, V. Savona, et al., *Phys. Rev. Lett.* **103**, 207403 (pages 4) (2009), URL <http://link.aps.org/abstract/PRL/v103/e207403>.
 - [5] A. Majumdar, A. Papageorge, E. D. Kim, M. Bajcsy, H. Kim, P. Petroff, and J. Vučković, *Phys. Rev. B* **84**, 085310 (2011).
 - [6] R. Bose, D. Sridharan, G. S. Solomon, and E. Waks, *Appl. Phys. Lett.* **98**, 121109 (2011).
 - [7] A. Laucht, J. M. Villas-Bôas, S. Stobbe, N. Hauke, F. Hofbauer, G. Böhm, P. Lodahl, M.-C. Amann, M. Kaniber, and J. J. Finley, *Phys. Rev. B* **82**, 075305 (2010), URL <http://link.aps.org/doi/10.1103/PhysRevB.82.075305>.
 - [8] K. J. Xu and C. Piermarocchi, *Phys. Rev. B* **84**, 115316 (2011), URL <http://link.aps.org/doi/10.1103/PhysRevB.84.115316>.
 - [9] E. B. Flagg, J. W. Robertson, S. Founta, W. Ma, M. Xiao, G. J. Salamo, and C.-K. Shih, *Phys. Rev. Lett.* **102**, 097402 (2009), URL <http://link.aps.org/doi/10.1103/PhysRevLett.102.097402>.
 - [10] A. Majumdar, E. Kim, Y. Gong, M. Bajcsy, and J. Vučković, *Phys. Rev. B* **84** (2011).
 - [11] C. Roy and S. Hughes, *Phys. Rev. Lett.* **106**, 247403 (2011).
 - [12] S. M. Ulrich, S. Ates, S. Reitzenstein, A. Löffler, A. Forchel, and P. Michler, *Phys. Rev. Lett.* **106**, 247402 (2011).
 - [13] D. Englund, A. Faraon, I. Fushman, N. Stoltz, P. Petroff, and J. Vučković, *Nature* **450**, 857 (2007).
 - [14] D. Englund, A. Majumdar, M. Bajcsy, A. Faraon, P. Petroff, and J. Vučković, arXiv:1107.2956 (2011).
 - [15] A. Majumdar, A. Faraon, E. D. Kim, D. Englund, H. Kim, P. Petroff, and J. Vučković, *Phys. Rev. B* **82**, 045306 (2010).

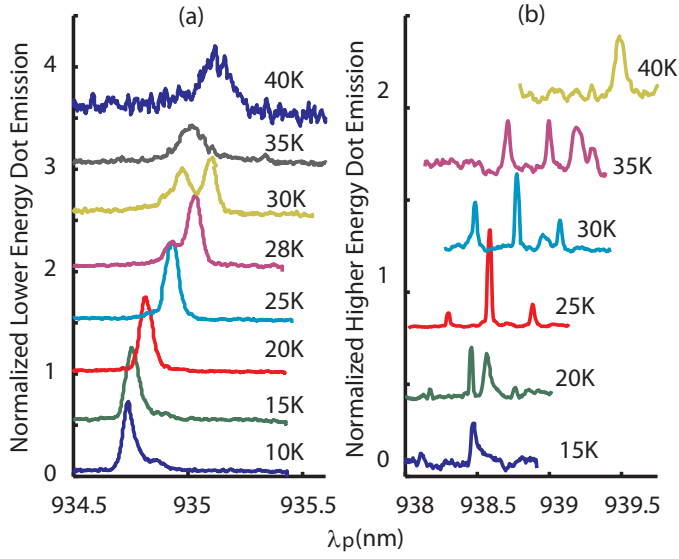


FIG. 6: (color online) Effects of temperature on dot to dot coupling and the resulting frequency conversion of the pump. In the experiments, the wavelength of the pump, λ_p , is scanned through one QD and the peak intensity of the other QD is monitored. In (a), the pump is scanned through the higher energy QD and the down-converted light from the lower energy QD is collected and plotted, while in (b), we plot the up-converted light emitted from the higher energy QD as the pump is scanned through the lower energy QD. In both figures, the plots are vertically offset for clarity.